Reversibility and Irreversibility in Spin-Glasses: The Free-Energy Surface

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The free-energy surface of a mean-field model for a spin-glass in a field H is studied numerically. Field-cooled (FC) and zero-FC magnetizations are obtained that look qualitatively like experiment. The FC state is shown to satisfy Maxwell's relations. Changing H below T_g generally leads to irreversible behavior. In the absence of relaxation effects, it is found that all processes obtained upon cooling are reversible. New hysteresis effects are predicted.

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We have gained insight into some aspects of the theory of spin-glasses from Monte Carlo simulations.[1] Recent theoretical ideas[2] also look promising. However, none of these approaches lends itself to establishing a simple physical picture. What is lacking is a detailed study of the complicated free-energy surface, F, which one can use to begin to understand in more physical terms the reversible and irreversible behavior, and thus gain insight into the strong history dependence of all measurements in these systems. The obvious space in which to study F is a function of the average local spin variables $\{m_i\}$. Thouless, Andersen, and Palmer[3] (TAP) have computed $F^{\mathrm{TAP}}[m_i]$ for the infinite-range Ising model. Ideally one should explore the behavior of F^{TAP} as the magnetic field H and temperature T are changed. It appears, however, that solutions to the minimization condition $\partial F^{\mathrm{TAP}}/\partial m_i = 0$ require the use of sophisticated iterative techniques[4] which involve matrix algebra. This imposes a limit on the number of particles N. Furthermore, there are unphysical minima to which the system will easily flow.

To determine whether this scheme is promising and to gain immediate insight into the behavior of F in spin-glasses, we have numerically studied the much simpler mean-field free-energy surface for Ising spins ($g\mu_B \equiv 1$):

$$F = -\sum_{i<j} J_{ij} m_i m_j + (T/2)\sum_i \{(1+m_i)\ln[\tfrac{1}{2}(1+m_i)] + (1+m_i)\ln[\tfrac{1}{2}(1-m_i)]\} - H\sum_i m_i, \tag{1}$$

where J_{ij} is the nearest-neighbor exchange constant distributed according to

$$P(J_{ij}) = (2\pi)^{-1/2} \tilde{J}^{-1} \exp(-J_{ij}^2/2\tilde{J}^2)$$

and

$$-1 \leq m_i \leq 1.$$

We will show here that this model, while clearly oversimplified,[5] nevertheless yields a number of results which are in qualitative agreement with experiment. Furthermore, it makes predictions which can be checked. While fluctuation effects have not yet been included they can be superposed onto the present scheme, once the behavior of F is understood. Although our approach is numerical it is a step closer to analytical theories than Monte Carlo simulations, for it builds in the broken symmetry $m_i \neq 0$ and it allows us to check such things as the Maxwell relations (MR) which are the subject of current controversy.[6] In addition, we can readily study the degree to which two minima are correlated at any temperature and thereby deduce barrier heights.[7]

In the present work we adopt the viewpoint that spin-glasses should be viewed as nonergodic systems so that all degenerate ground states are not equally important. Rather, the focus is on the appropriate field-cooled state and its associated accessible minima. This approach is to be contrasted with thermodynamic calculations. It seems plausible that the difficulties[8] encountered in studying the spin-glass phase transition within a thermodynamic approach may be due to nonergodicity.

Our system consisted of several 30×30 and 100×100 square lattices and one $10\times10\times10$ three-dimensional (3D) system. We studied how a minimum of F evolved with H and T by using an iterative approach, which always generates minima.[9] For each new (T, H) we started our iterations at the $\{m_i\}$ corresponding to the minimum of F evaluated at the limit values for the previous T or H. We then changed the $\{m_i\}$ by using a random (updated) sequencing of the sites i until we got convergence at the nth iteration defined by

$$\sum_i (m_i^n - m_i^{n-1})^2 / \sum_i (m_i^n)^2 \leq 10^{-12}.$$

For definiteness, the spin-glass transition temperature T_g is defined by the lowest T at which $\sum_i m_i^2 = 0$, obtained by extrapolating to $N\to\infty$. For the 2D case we found $T_g = 3.4\bar{J}$ (here $k_B \equiv 1$). We organize our conclusions as follows.

(1) *Changes in T.*—Any minimum which exists at temperature T_0 was found to also exist at all $T<T_0$. However, if we started with a random minimum and heated, the minimum disappeared and the system found its way to a nearby state. This irreversibility upon heating was clearly manifested for small N by the appearance of "jumps" in the thermodynamic variables. For large N we saw it by noting that cooling (after heating) always led to a different state than we started with.

(2) *Changes in H.*—We found that changes in H were always irreversible for $H<H_R(T)$. In our finite-size systems $H_R=0$ for $T\geq 1.2T_g$, so that above this T there is a single minimum in F; for $T=0.35T_g$, $H_R=0.94T_g$. Irreversibility arises because as H is increased or decreased, minima disappear and the system finds its way to a nearby state. Thus, unlike changes in T, minima continuously appear as well as disappear as H is varied at fixed T. Despite conjectures[10] to the contrary, it does not appear that below T_g there is a nonzero critical field below which changes in H are reversible.

(3) *Field-cooled (FC) state.*—A unique FC state is always obtained. It is insensitive to the sequencing of the $\{m_i\}$ changes, provided one starts cooling at $T>1.2T_g$. This result was seen even in extremely weak fields. For each (H, T) this state has the lowest free energy of any state we generated. However, we did not make a systematic search. In agreement with experiment the FC magnetization is totally reversible in T.[11]

(4) *Zero-field-cooled (ZFC) state.*—The states obtained by cooling in zero field were not unique, possibly because of finite-size effects. They varied depending on our choice of sequencing the changes in $\{m_i\}$. It follows from conclusion (1) that the system would not lose any minimum (obtained by zero-field cooling) at a lower T, so that in the absence of relaxation effects each ZFC state is as reversible as any finite-field-cooled state. However, a measurement of the magnetization of such a ZFC state necessitates applying H below T_g and [from conclusion (2)] leads to irreversibility.

(5) *High-field demagnetized state.*—We sought to construct a new state by first cooling in zero field, next applying $H>H_R(T)$, and then decreasing H to some final value $H_F<H_R$. For $T=0.35T_g$ the state so obtained is a rather deep minimum for all but small values of $H_F<0.14T_g$. The corresponding values of F and M [see Fig. 2(d)] are a few percent higher than those obtained by field cooling. The state is highly correlated with the FC state. Thus it appears that such a high-field demagnetized state will rapidly decay into the FC state. These predictions can be checked experimentally.

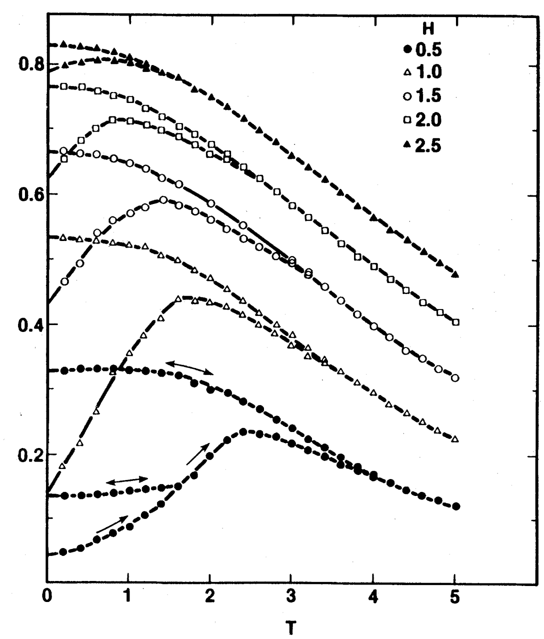

FIG. 1. Field-cooled (upper curves) and zero-field-cooled (lower curves) magnetization vs temperature $T/\bar{J}$ for various $H/\bar{J}$.

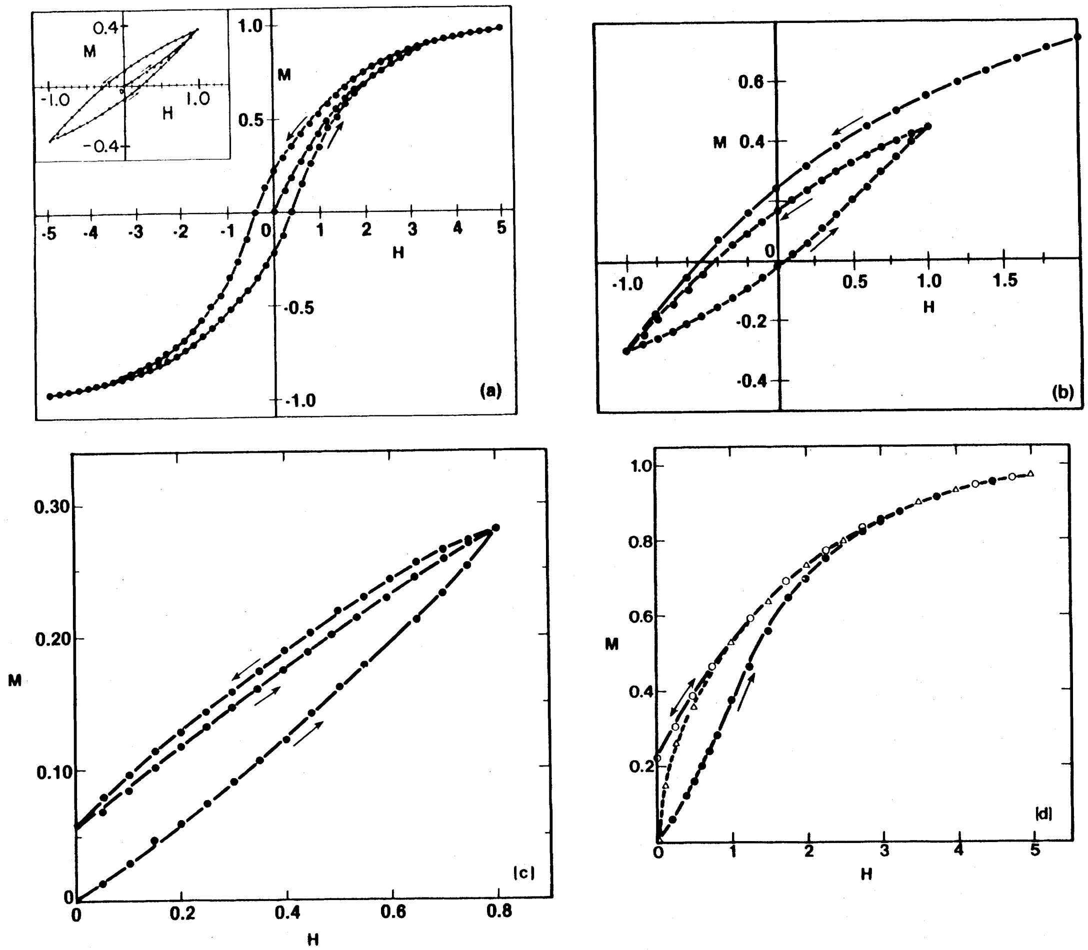

FIG. 2. Magnetization vs H at $T = 0.35T_g$ for (a) symmetric field sweeps for high and low (inset) maximum H, (b) demagnetized (from high field) loop, (c) positive-H minor loop, and (d) positive-H minor "loop" (open circles). Field-cooled magnetization is shown by triangles. Here $N = 10^4$.

(6) *Thermodynamic relations.*—We have checked the thermodynamic relations $S = (-\partial F/\partial T)_H$ and $M = (-\partial F/\partial H)_T$ in all the states we generated by evaluating the derivatives numerically and comparing the resulting quantities with M and S calculated directly. We found that these equalities were violated in all cases except for the FC state. This violation comes about because of irreversibility effects, i.e., hopping between minima. It is straightforward to see that in the FC state all changes in H and T are reversible: Changing H far above T_g leads to no hysteresis and all states obtained upon cooling are reversible. Our result that the MR are satisfied within the FC state is in agreement with some[12] but not all previous[6] claims. That they are violated in all other states is intimately related to the breakdown of linear response theory in the non-FC states.

In Fig. 1 we plot the temperature dependence of the magnetization M^{FC} and M^{ZFC} obtained by FC (upper curves) and ZFC (lower curves) processes for a range of H values. These curves all correspond to a single 30×30 configuration. We have verified that they are qualitatively the same for other 30×30 and 100×100 configurations. However, for the 3D case we found that the FC and ZFC curves intersected closer to $T^{\max}(H)$ than in 2D. These results look qualitatively similar to experiment in that at low T, M^{FC} is nearly constant, whereas M^{ZFC} has a maximum at some $T = T^{\max}(H)$ which approaches T_g as $H \to 0$. These results differ from experiment in ways that can be readily attributed to finite-size effects. We see a broader maximum in M^{ZFC} and a lower T at which M^{FC} saturates due to the "rounding" of the transition. This leads to a meeting of the two curves (for the same H) at a temperature higher than the maximum in M^{ZFC}. In fact, these curves are in strikingly good agreement with measurements[13] made on nonannealed spin-glasses in which it is argued that inhomogeneities give rise to a range of T_g's (presumably, much as do finite-size effects).

In the lowest ZFC magnetization curve in Fig. 1 we have illustrated what happens if below T^{max} the temperature is decreased down to zero and then increased again. The magnetization thus obtained is roughly constant in temperature and reversible. This phenomenon has been seen in recent experiments.[11]

We have studied in detail the hysteresis curves obtained for a number of situations (at $T=0.35T_g$). The width of the hysteresis loop has been used[14] as a measure of the anisotropy energy of a spin-glass, which anisotropy also plays a role in spin resonance experiments.[14] We emphasize here that the hysteretic behavior we observe comes entirely from the behavior of the free energy surface. It derives from the fact that minima disappear and new ones appear as H is varied so that the system wanders from one state to another. Figure 2(a) shows the hysteresis loop obtained after zero-field cooling by applying a maximum field $H>H_R$. The inset corresponds to the case of a small maximum field. Both loops are symmetric and in qualitative agreement with what is observed in Monte Carlo simulations[15] and *Au*Fe experiments.[16] If the system is started from a large magnetic field $H=H_R$ after which the field is then decreased to a small negative value and the loop then closed, we see a displaced hysteresis loop, as is shown in Fig. 2(b) and seen[16] in field-cooled *Au*Fe spin-glasses. In Fig. 2(c) we show a minor hysteresis loop for $H>0$; in this case the field is first increased from zero to some maximum value, then decreased to $H=0$, and then increased back up to the maximum value. The larger the maximum H is, the smaller the area contained in the loop. To illustrate this, in Fig. 2(d) we show the results for a field sweep up to the reversible field H_R. Here the "loop" has collapsed to a single line on the scale of the figure. We plot the FC magnetization at this T for comparison purposes.

We conclude by noting that the main contributions of the present work are (1) to suggest an explanation for the physical origin of anisotropy (as seen in hysteresis data); (2) to show how to understand history-dependent measurements, which *cannot* be understood in the context of analytic configuration-averaged or thermodynamic calculations; (3) to make predictions for other hysteretic behavior [see Figs. 2(b)-2(d)]; (4) to illustrate clearly how irreversibility arises from changing H and T; and, thereby, (5) to show that the field-cooled state alone satisfies thermodynamic identities such as the Maxwell relations.

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